



US007674916B2

(12) **United States Patent**
Werpy et al.

(10) **Patent No.:** US 7,674,916 B2
(45) **Date of Patent:** Mar. 9, 2010

(54) **PROCESS FOR PRODUCING CYCLIC COMPOUNDS**

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(*) Notice: Subject to any disclaimer, the term of this
patent is extended or adjusted under 35
U.S.C. 154(b) by 277 days.

(21) Appl. No.: **11/688,022**

(22) Filed: **Mar. 19, 2007**

(65) **Prior Publication Data**

US 2007/0173643 A1 Jul. 26, 2007

Related U.S. Application Data

(62) Division of application No. 10/731,108, filed on Dec.
10, 2003, now Pat. No. 7,199,250.

(60) Provisional application No. 60/435,469, filed on Dec.
20, 2002.

(51) **Int. Cl.**
C07D 207/267 (2006.01)
C07D 213/63 (2006.01)

(52) **U.S. Cl.** **548/552**; 546/290

(58) **Field of Classification Search** 548/552;
546/290

See application file for complete search history.

(56) **References Cited**

U.S. PATENT DOCUMENTS

2,267,757 A 12/1941 Schuster et al.
2,674,602 A 4/1954 Schuster
3,080,377 A 3/1963 Liao
3,092,638 A 6/1963 Liao et al.
3,109,005 A 10/1963 Lidov
3,198,808 A 8/1965 Walldorf et al.
3,448,118 A 6/1969 Chichery et al.
3,634,346 A 1/1972 McKeon et al.
3,637,743 A 1/1972 Prince
3,661,894 A 5/1972 Wehner et al.
3,681,387 A 8/1972 Hollstein et al.
3,767,644 A 10/1973 Fukuoka et al.
3,775,431 A 11/1973 Rodewald
3,812,148 A 5/1974 Hollstein et al.
3,884,936 A 5/1975 Hollstein
3,966,763 A 6/1976 Greene
4,012,418 A 3/1977 Schaafsma et al.
4,151,171 A 4/1979 Kurkov
4,152,331 A 5/1979 Meijer et al.
4,263,175 A 4/1981 Pesa et al.
4,356,124 A 10/1982 Pesa et al.
4,404,391 A 9/1983 Meyer et al.
4,420,620 A 12/1983 Murib
4,731,454 A 3/1988 Otake et al.

4,780,547 A 10/1988 zur Hausen et al.
4,800,227 A 1/1989 Matson
4,814,464 A 3/1989 Olsen
4,841,069 A 6/1989 Olsen
4,851,546 A 7/1989 Graham et al.
4,885,371 A 12/1989 Tracy et al.
4,904,804 A 2/1990 Matson
5,101,044 A 3/1992 Schuster et al.
5,101,045 A 3/1992 Koehler et al.
5,106,730 A 4/1992 Van Ness et al.
5,157,127 A 10/1992 Weyer et al.
5,276,165 A 1/1994 Weyer et al.
5,338,861 A 8/1994 Botta et al.
5,347,021 A 9/1994 Taylor et al.
5,401,856 A 3/1995 Grey et al.
5,434,273 A 7/1995 Weyer et al.
5,478,950 A 12/1995 Bergfeld et al.
5,508,396 A 4/1996 Steffen

(Continued)

FOREIGN PATENT DOCUMENTS

CN 1104635 7/1995

(Continued)

OTHER PUBLICATIONS

Olsen's, 1989, CAS: 111: 232565.*

(Continued)

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(57) **ABSTRACT**

The invention includes methods of processing an initial di-carbonyl compound by conversion to a cyclic compound. The cyclic compound is reacted with an alkylating agent to form a derivative having an alkylated ring nitrogen. The invention encompasses a method of producing an N-alkyl product. Ammonia content of a solution is adjusted to produce a ratio of ammonia to di-carboxylate compound of from about 1:1 to about 1.5:1. An alkylating agent is added and the initial compound is alkylated and cyclized. The invention includes methods of making N-methyl pyrrolidinone (NMP). Aqueous ammonia and succinate is introduced into a vessel and ammonia is adjusted to provide a ratio of ammonia to succinate of less than 2:1. A methylating agent is reacted with succinate at a temperature of from greater than 100° C. to about 400° C. to produce N-methyl succinimide which is purified and hydro-genated to form NMP.

15 Claims, 2 Drawing Sheets